Performance of new infrared beamline U12IR at the National Synchrotron Light Source

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The instrumentation and performance of the new infrared beamline U12IR at the National Synchrotron Light Source of Brookhaven National Laboratory is described. This beamline utilizes infrared synchrotron radiation from a bending magnet. A combination of beamline design features and spectroscopic instrumentation allows the facility to reach the extremely low frequency limit of $\sim 2\ \mathrm{cm}^{-1}$ (i.e., 60 GHz or a photon energy of 250 μeV). The infrared light from the synchrotron emission at U12IR is compared to standard thermal sources and reveals substantial benefits for the study of small samples. In particular, the intensity of the synchrotron radiation in the far infrared can be as much as 200 times greater than that from a blackbody when millimeter-sized samples are measured. The effects of diffraction and noise on beamline performance are also discussed.
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I. INTRODUCTION

Synchrotron radiation facilities were developed mainly to produce photons in the vacuum ultraviolet through x rays for which alternative sources are inadequate. Later it was recognized that useful light is produced far into the infrared. As an infrared source, it possesses several advantages over conventional blackbody sources. Among the most important is its brightness advantage, which can approach 1000 times that of thermal sources. The synchrotron source is pulsed on a subnanosecond time scale, and spans the full optical range: microwave through ultraviolet. Because the emitted power depends in a known way on the electron beam current in the storage ring—a quantity that can be measured very accurately—the synchrotron is an absolute source for the infrared.[1] Infrared synchrotron radiation does suffer several drawbacks when compared to thermal sources. Except for the very longest wavelengths, the overall power of the synchrotron is typically smaller than that from thermal sources.[2,3] Also, synchrotron radiation is available only at major scientific laboratories, and requires specialized vacuum and optical instrumentation to extract and deliver the light to a spectrometer.[4,5] Finally, the characteristic noise differs from conventional sources.

Nevertheless, several experiments in the infrared benefit from the use of the synchrotron infrared light. Surface spectroscopy,[1,6] (IR) microscopy,[7] high pressure studies,[8] far-infrared ellipsometry,[9] and broadband pump–probe experiments[10] are some examples of measurements techniques that exploit the intrinsic characteristics of infrared synchrotron radiation.

Since the mid-1980's the National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory has developed and maintained an infrared capability. U4IR[11] was the first infrared beamline at the NSLS and, with increasing demand for measurement time, has been followed by a series of new infrared ports presently under construction and commissioning. This also allowed for some specialization, and the new beamline U12IR has been designed specifically for optimal performance in the far infrared. The beamline will also be used for time-resolved pump–probe spectroscopy in conjunction with a synchronized pulsed laser system.[12–14] However, there are no particular beamline design features for this purpose. In this article we present details of the U12IR beamline and characterize its performance.

II. BEAMLINE LAYOUT

In contrast to bending-magnet beamlines for vacuum ultraviolet (VUV) or x rays, an infrared beamline requires large extraction optics, due to the increasingly larger angles into which synchrotron radiation is emitted as wavelengths increase. This angle is given by[15,16]

$$\theta = 1.6\left(\frac{\lambda}{\rho}\right)^{1/3}, \tag{1}$$

where ρ is the electron orbit bending radius and λ is the wavelength. Thus, efficient extraction of the infrared from a synchrotron is increasingly difficult towards longer wavelengths. U12IR is a conventional bending magnet infrared beamline, built to collect a solid angle of 90 mrad×90

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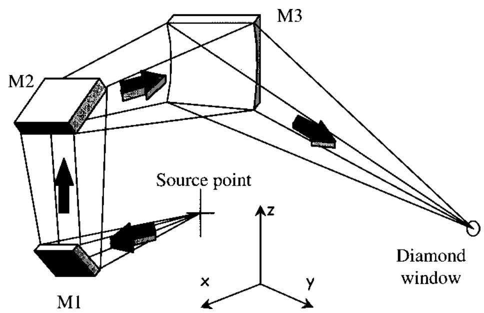

FIG. 1. A schematic of the extraction mirrors for U12IR. M1 is a gold-coated, water cooled, SiC mirror, placed close to the electron beam. This mirror subtends a 90 mrad×90 mrad solid angle, collecting all the light emitted down to approximately 30 cm^{-1}. M2 and M3 are Al-coated Pyrex and M3 is an ellipsoid that images the source point at the diamond window.

mrad.[17] The NSLS-VUV ring has a bending radius of 1.91 m, therefore the beamline collects (per horizontal angle) 100% of the infrared down to 30 cm^{-1}, falling to abut 40% at 2 cm^{-1}.[18]

Details of the beamline's optical, vacuum, and protection systems are presented elsewhere.[17] Here we review just a few of the optical components. The beamline can be viewed as two parts: an ultrahigh vacuum (UHV) section, directly connected to the ring, containing the extraction mirrors; and a rough vacuum section, containing optics for transferring the light to the spectrometer end stations. The two sections are separated by an 11 mm diam wedged chemical vapor deposited (CVD) diamond window. The infrared light is extracted vertically from the ring by a set of three mirrors, shown schematically in Fig. 1. M1 and M2 are plane mirrors, while M3 is an ellipsoidal mirror that focuses the synchrotron emission to a point just beyond the diamond window. The calculated power delivered through the diamond window is shown in Fig. 2. The drop near 15 000 cm^{-1} is due to the gold coating of M1 and the small absorption at 12 500 cm^{-1} is due to the aluminum coating of M2 and M3.

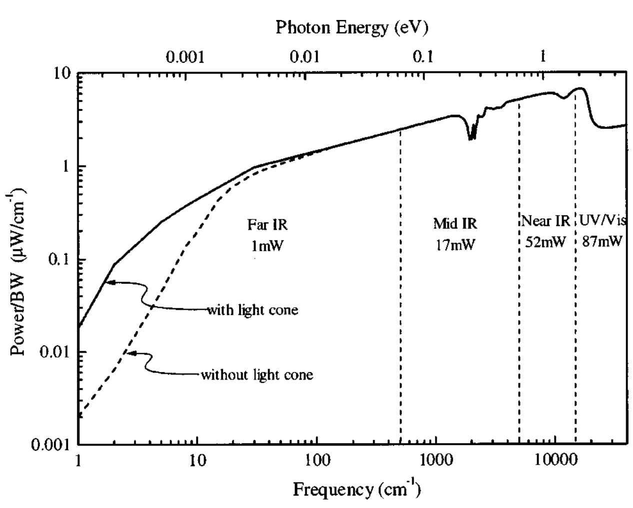

FIG. 2. Power transmitted by the diamond window at the U12IR beamline. The absorption around 2600 cm^{-1} is due to two-phonon absorption in the diamond window. The band at 12 500 cm^{-1} is due to electronic transitions on the Al mirrors. The edge at ~15 000 cm^{-1} is due to absorption in the gold coating of the first mirror. The band gap of the diamond window is responsible for a cutoff at ~40 000 cm^{-1}.

Vibrational absorption in the diamond window is responsible for the structure around 2700 cm^{-1} and its above-band gap absorption gives the strong cutoff at 40 000 cm^{-1}. The maximum power passing through the diamond window, integrated from far infrared to near UV, is 170 mW.

Since the geometric source size is smaller than the 11 mm diam diamond window, most of the light collected by the extraction mirrors system passes directly through. But at the longest wavelengths, diffraction causes the focused spot size to increase. To minimize the losses due to diffraction one can, in principle, use very large optics. However, this necessitates having adequate space as well as a large window. Diamond is the only material that is relatively transparent from the far infrared to the near ultraviolet, and large diamond windows are not practical. The collection of this long wavelength light is aided by a ''light cone'' just upstream of the diamond window. This cone has gold-coated interior walls (for improved IR efficiency) that slope at a 6° angle (relative to the central axis). It tapers from a 62 mm diam entrance aperture down to match the 11 mm aperture at the diamond window. With our ~f/12 light collection geometry, the diameter of the light spot (central Airy disk) at the diamond window, due to diffraction, is approximately 30 λ (λ being the wavelength). Therefore, part of the light below 30 cm^{-1} will hit the walls of the cone at grazing incidence and will be redirected toward the diamond window. Depending on the number of reflections, the light exits the cone at angles approaching 24° (400 mrad). Based on the ratio between the areas of the diffraction limited spot size and the diamond window, we can conclude that the optical cone improves the optical efficiency by a factor of ~4 at 7 cm^{-1}. This factor saturates around a value of 35 below 4 cm^{-1} as the diffraction pattern overfills even the entrance of the cone. The overall beamline schematic, including the optical cone, is shown in Fig. 3.

After passing through the diamond window the light leaves the UHV section and enters the rough vacuum section. From here it is sent to one of two available end stations at U12IR: a lamellar grating interferometer or a Bruker IFS 113v spectrometer. For the Bruker IFS 113v, the light leaving the diamond window is collimated by an off-axis parabolic mirror, transported to the spectrometer by plane mirrors and refocused at the entrance of the interferometer. The parabolic mirror is designed to collect ~90 mrad, i.e., it is matched to the UHV optics without consideration for the light cone. Therefore, the long wavelength benefits of the light cone are not realized for the Bruker. Additional diffraction losses occur in the transport optics such that the efficiency is further reduced below 100 cm^{-1}.

The far infrared is delivered to the lamellar grating instrument by a 1.27 cm diam brass light pipe. The light pipe begins immediately after the diamond window, and collects effectively all the infrared exiting from the diamond window aperture, including the light that reflects from the cone. The light pipe is useful for short distances and long wavelengths (beyond 10 μm), so it is well matched to the spectral range of the lamellar grating interferometer (100 μm and longer).

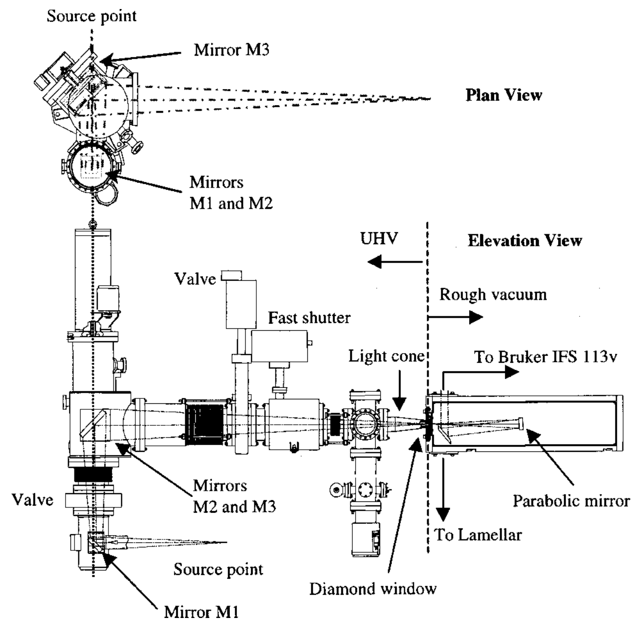

FIG. 3. The U12R UHV section. The light cone placed just before the diamond window improves the very far infrared (below 15 cm^{-1}) intensity. The mirror box after the diamond window can be configured to send the light to the Bruker IFS-113v interferometer or to the Lamellar grating interferometer. The parabolic mirror collimates the light that is transported by mirrors to the Bruker. The light is fed to the lamellar by 1/2 in. diam brass light pipes.

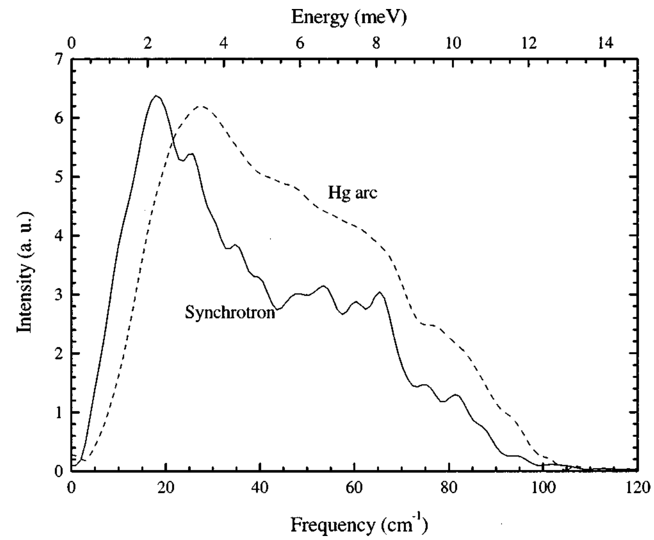

FIG. 4. Synchrotron (solid line) at 600 mA and Hg arc lamp (dashed line) spectra obtained with the Lamellar grating spectrometer. The transport of the light via light pipes makes the synchrotron lose its brightness advantage with respect to the thermal sources. In this spectrometer, the synchrotron radiation provides more light than the thermal sources below 20 cm^{-1}.

III. SPECTROMETERS AND BEAMLINE PERFORMANCE

The lamellar grating spectrometer[19] covers the 2–100 cm^{-1} (0.2–12 meV), long wavelength range. The instrument can be used with both the synchrotron source as well as its internal, high pressure Hg lamp (a GE UA-3 operating at 250–300 W). For the synchrotron, the light is introduced through 2 m of light pipe, which includes two right-angle bends. The measured efficiency of this length is about 50%. Spectra obtained with both the Hg arc lamp and the synchrotron source are shown in Fig. 4. The synchrotron spectrum is for a ring current of 600 mA,[20] an average current for the VUV ring at NSLS. We note that the two sources provide a comparable signal at the detector, although the synchrotron outperforms the Hg lamp for frequencies below 25 cm^{-1}. At 5 cm^{-1} the synchrotron advantage approaches a factor of 5. The crossover point where the synchrotron outperforms the thermal source is substantially lower than that observed by other groups. This is due to the differences in temperature and size for the thermal source as well as the collection optics. A typical IR spectrometer uses a globar or Hg lamp with dimensions of a few mm. Source collection optics have f numbers between $f/3$ and $f/4$. The lamellar grating interferometer's source is large and situated in a cylindrical cavity which serves to increase the effective f number to $f/1$ or faster. This, along with the additional light pipe used for transporting the synchrotron infrared, accounts for the crossover occurring below 100 cm^{-1}. It is worth noting that the light cone and subsequent light pipe, although efficient for overcoming diffraction losses, can lead to reduced source brightness from multiple reflections and ''mode mixing.'' Still, the beamline does allow spectra to be collected to lower frequencies (e.g., 3 cm^{-1} using a 1.2 K bolometer), and more importantly, maintains the pulsed nature of the synchrotron source for use in pump–probe experiments. This type of experiment is described elsewhere.[14]

The Bruker 113v interferometer covers the spectral range from 10 to 8000 cm^{-1} (1 meV–1 eV). In contrast to the lamellar interferometer, the optical system for this spectrometer, is designed to maintain the synchrotron's high brightness. The brightness of the source is defined as the power (or photon flux) per source emittance (i.e., the product of the source area and solid angle into which the source emits). When the source's emittance exceeds the throughput of the optical system, the source brightness rather than source power determines the signal that reaches the detector. An example of a low throughput optical system is one with a small aperture, such as that which naturally occurs when performing spectroscopy on very small specimens (microspectroscopy). We illustrate the brightness advantage of the synchrotron source by collecting spectra through small apertures situated at the sample location of the IFS 113. Spectra were collected using both the synchrotron source and the spectrometer's internal thermal sources (globar for frequencies above 400 cm^{-1}, and a high pressure Hg arc lamp for frequencies below 400 cm^{-1}). The relative signal levels for synchrotron compared to thermal source are shown in Fig. 5(a) with a variety of apertures placed at the sample focal point. As the aperture size decreases, the ratio increases, reaching in excess of 100 for the 0.5 mm aperture. Even for a large aperture, the thermal source outperforms the synchrotron only for frequencies above 500 cm^{-1}. Figure 5(b) shows the integrated intensity of the light transmitted through the apertures as a function of the aperture size. The frequency range used for the integration is 10–100 cm^{-1}; similar results are obtained in other frequency ranges. The integrated intensity is normalized by the value obtained for the largest aperture (10 mm) for each source. Whereas the

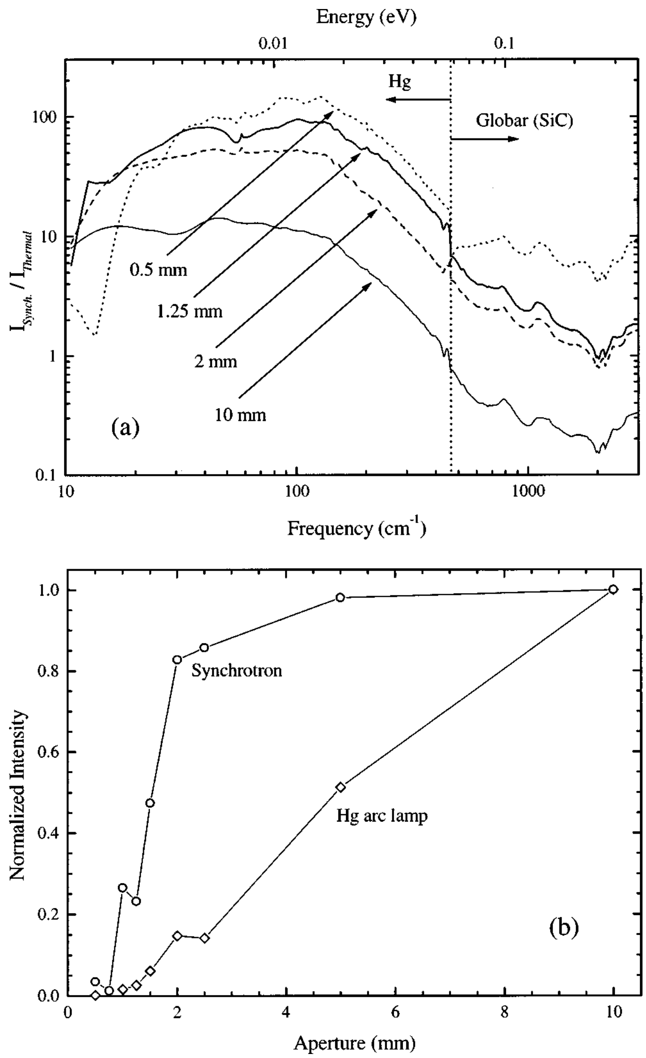

FIG. 5. (a) Ratio of the intensity from synchrotron radiation I_s and thermal sources I_{th} in the NSLS U12IR beamline using a Bruker IFS-113v spectrometer. A mercury arc lamp is used as thermal source below 450 cm^{-1}, whereas a globar (SiC) is used above. The step observed in the ratio when changing sources can be attributed to their different effective blackbody temperatures. (b) Integrated intensity in the far infrared region (0–100 cm^{-1}) as a function of the size of an aperture placed at the sample position for the synchrotron and Hg arc sources. Each curve is normalized by the integrated intensity for the 10 mm aperture. There is a monotonic increase in the integrated intensity for the thermal source, whereas the synchrotron light is unaffected by aperture size changes down to a 2 mm spot. The observed behavior for the other spectral ranges is similar.

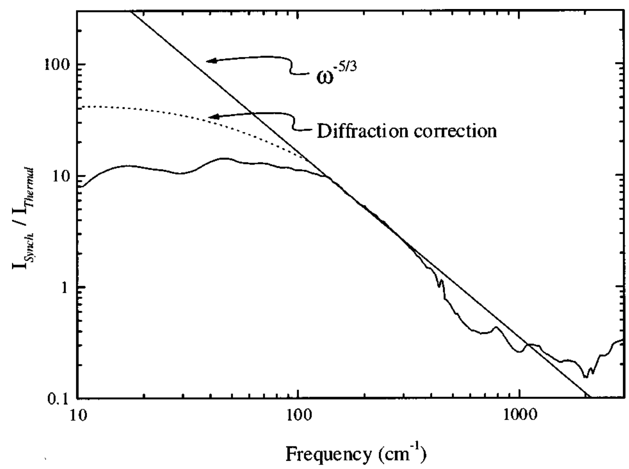

FIG. 6. Ratio between synchrotron and thermal sources I_s/I_{th} for the 10 mm aperture compared to the expected value from synchrotron and blackbody power calculations. The solid straight line is the expected $\omega^{-5/3}$ ratio. The dotted line takes into account the diffraction due to diffraction effects in the beamline.

increase in the integrated intensity for the thermal sources goes almost monotonically with the aperture size, the synchrotron light only begins to fall off below a 2 mm size aperture. This result provides a strong indication that over 80% of the far-infrared synchrotron radiation in the U12IR beamline Bruker interferometer is concentrated in a 2 mm size spot.

IV. DISCUSSION

Comparison of the synchrotron and thermal sources must take into account the different wavelength dependences of the radiation from each. At long wavelengths ($\lambda \gg \lambda_c$), (λ_c being the synchrotron cutoff wavelength, typically in the x-ray region) the synchrotron radiation intensity (power/source-area/bandwidth) is proportional to $\lambda^{-7/3}$.[21] The radiation emitted by a blackbody at wavelengths much larger than hc/kT is proportional to λ^{-4}. Thus the ratio of synchrotron radiation intensity to thermal-source intensity should be proportional to $\lambda^{5/3}$, or equivalently to $\omega^{-5/3}$.[22] In the NSLS-VUV ring the cutoff wavelength is 20 Å and the above approximation is valid in the entire infrared range. However, a typical Hg arc lamp has an effective blackbody temperature around 2000 K ($hc/kT \sim 7$ μm=1400 cm^{-1}) and a typical globar 1200 K has $hc/kT \sim 12$ μm=830 cm^{-1}. Because our blackbody measurements use a mercury arc lamp below 450 cm^{-1} and a globar at higher wave numbers, the above approximation holds only for the arc lamp spectra.

Figure 6 shows the ratio measured for the largest aperture (10 mm). This aperture size collects, in practice, all the light from both the synchrotron and the thermal sources. Therefore it is a good representation of the total power emitted by these sources. The solid straight line is the $\omega^{-5/3}$ expected behavior. The ratio between the two sources follows the $\omega^{-5/3}$ law in the 150–450 cm^{-1} range. Above 450 cm^{-1}, as discussed above, the long-wavelength approximation to the blackbody is no longer valid. The downward step in the ratio of Fig. 5(a), when passing from the Hg arc lamp to the globar, can be attributed to the difference in the effective temperature of both thermal sources and to absorption due to the quartz envelope of the Hg arc lamp.

Below 150 cm^{-1} we attribute the deviations to diffraction effects. In contrast to the thermal sources, which are located inside the Bruker 113v spectrometer, the synchrotron light is transported 7 m from the diamond window to a spherical focusing mirror inside the spectrometer. The parabolic mirror that collimates the light has a 5×5 cm^2 cross section. Even though the light is collimated, after reflection on the parabolic mirror, diffraction of the light produces a divergence angle given by

$$d \sin\theta \approx \lambda, \tag{2}$$

where d is the parabolic mirror size and λ is the wavelength

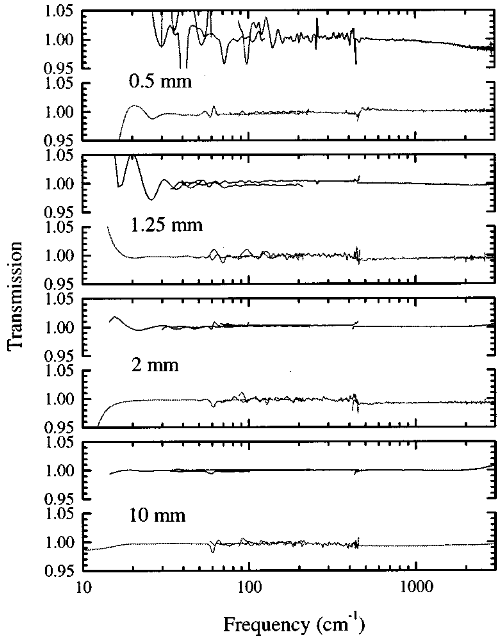

FIG. 7. 100% lines for synchrotron and thermal sources in the Bruker IFS-113v interferometer. In all panels, the upper curve is for thermal sources and the lower for synchrotron radiation. Due to the small spot size of the synchrotron source, the effect of aperture (sample) size on the measurement noise is negligible. In contrast, for the thermal sources, the noise increases rapidly for smaller apertures. Beam motion introduces an aperture-independent noise into the synchrotron measurements.

of the light. The spherical mirror inside the Bruker interferometer has a 7.6 cm cross section and it is located 6.5 m after the parabolic mirror. Considering that the light completely fills the parabolic mirror,[23] diffraction effects will make the spot at the last mirror have an apparent size of

$$l=d+\frac{2L\lambda}{d}, \tag{3}$$

where L is the distance between the parabolic mirror and the last mirror. We used a small-angle approximation ($\sin\theta \sim\theta$) to obtain Eq. (3). If l is larger than the dimension l' of the last mirror, the light will overfill this mirror and the lost intensity will be approximately proportional to the difference in the area of the spot and the mirror $[(l/l')^2;\ l>l']$. The dotted line in Fig. 6 takes into account the diffraction effect in the propagation of the light and describes qualitatively the loss of power in the synchrotron radiation at low energies. The discrepancies can be attributed to the other mirrors in the path between the parabolic collimating mirror and the spherical focusing one. Every mirror in between will act as an aperture and diminish the intensity at long wavelengths. Note that in Fig. 6, we also disregarded the effects below 30 cm^{-1} due to diffraction at the optical cone.

Figure 7 illustrates the signal to noise in the spectrum as the aperture size in the Bruker IFS 113v is varied. ''100% lines'' (the ratio of two successive spectra without a sample) are shown for both synchrotron and thermal sources in the far and mid-infrared ranges. Note that the noise in the measurements obtained with the synchrotron radiation remains almost independent of the aperture size down to 0.5 mm, in stark contrast with thermal sources. Using a Hg lamp, it is almost impossible to get reasonable data below 100 cm^{-1} for small samples without accumulating data for prohibitively long periods of time. Using the synchrotron, and with only 64 scans (about 1 min acquisition time), one can get a high signal-to-noise ratio on samples as small as 500 μm down to 20 cm^{-1} as shown on the top panel of Fig. 7. The limit of 20 cm^{-1} is related to diffraction effects from the 500 μm aperture. Finally, the noise that we observe in the fair infrared can be attributed to beam motion, either real (intrinsic) or apparent.[24] The NSLS VUV ring has various feedback systems to stabilize the beam position and minimize intrinsic low frequency motions. Beam noise at higher frequencies is attributed to the storage ring's rf accelerating system, and efforts continue toward minimizing their effects. Apparent beam motion occurs when optical elements between the spectrometer and synchrotron source are not perfectly fixed with respect to each other. In either case, the resulting noise scales with the signal. For high throughput experiments, the source noise can exceed the detector noise, and in this situation one achieves better signal-to-noise with the conventional (thermal) spectrometer source.

The characterization and operating capabilities of the new U12IR beamline at the NSLS at Brookhaven National Laboratory have been described. This beamline operates at the far infrared down to 2 cm^{-1}. Two interferometers are available, covering the ranges of 2–100 and 10–5000 cm^{-1}, respectively. The optical transport by light pipes in the low energy spectrometer diminishes the brightness of the synchrotron light somewhat with respect to conventional thermal sources, but the synchrotron light offers more intensity in the region below 20 cm^{-1}. In the far- and mid-infrared interferometer, where light pipes are not used, the spot size of the synchrotron light has a characteristic size of 2 mm. For small samples, the intensity gain of the synchrotron radiation reach values higher than 100 when compared to thermal sources. Due to the long path needed to transport the light from the storage ring to the spectrometer, diffraction effects limit the advantage of the synchrotron radiation in the very far infrared. Noise originating from beam motion and beamline vibrations make the infrared synchrotron radiation increasingly less suited for large ($>3\times3$ mm^2) samples.

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